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14. ABSTRACT The main objective of this contract is to provide support for a graduate student (M.S. Bhuiyan) from University of Houston to do research at Oak Ridge National Laboratory for a Ph.D. thesis. Mr. Bhuiyan finished all the requirements and examinations for a Ph.D. degree. He published three papers and is expected to defend his thesis in December 2004. His thesis is entitled "Study of Chemically Deposited Buffer Layers for Y B C O Coated Conductors".					
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F49620-02-1-03-17

**Epitaxial Growth of Solution Based Buffer Layers on Biaxially Textured Metal
Substrates for YBCO Coated Conductors**

Kamel Salama
University of Houston

Objectives: This program started on 7/1/2002. Its main objective is to provide support for a Ph.D. graduate student from the University of Houston (Shafiqur R. Bhuiyan) to do Ph.D. thesis research at Oak Ridge National Laboratory (ORNL). The research is aimed at developing a low-cost, non-vacuum method for growth of epitaxial buffer layers on biaxially textured Ni-and Cu- based substrates for YBCO coated conductors.

Status of effort: Starting 7/01/2002, Mr. Bhuiyan took 3 courses toward his Ph.D. degree and also passed the screening examination. In January 2003, Mr. Bhuiyan went to ORNL to work on his Ph.D. thesis research under the supervision of Dr. M. Parans Paranthaman of the Chemical Science Division, ORNL. In January 2004 he came back to University of Houston where he finished another three courses and passed the qualifying examination for the Ph.D. degree. In May 2004, Mr. Bhuiyan went back to Oak Ridge National Lab to continue working on his Ph.D. research; December 2004 is the target date for Bhuiyan to defend his Ph.D dissertation. A summary of his research effort as of now is outlined in the attached three documents. 1) A Paper published in the Journal of Superconductor Science and Technology entitled "MOD Approach for the Growth of Epitaxial CeO_2 Buffer Layers on Biaxially Textured Ni-W Substrates for YBCO Coated Conductors." 2) A paper entitled "Growth of Epitaxial Y_2O_3 film on Biaxially Textured Ni-W Substrates published in Mat. Res. Soc. Symposium Proc. Vol EXS-3, 2004. 3) A manuscript entitled "Epitaxial Growth of Solution Based Rare Earth Niobate, RE_3NbO_7 on Biaxially Textured Ni-W Substrates", to be submitted to Journal of Materials Research. Copies of these papers are attached.

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MOD approach for the growth of epitaxial CeO_2 buffer layers on biaxially textured Ni–W substrates for YBCO coated conductors

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Abstract

We have grown epitaxial CeO_2 buffer layers on biaxially textured Ni–W substrates for YBCO coated conductors using a newly developed metal organic decomposition (MOD) approach. Precursor solution of 0.25 M concentration was spin coated on short samples of Ni–3 at%W (Ni–W) substrates and heat-treated at 1100 °C in a gas mixture of Ar–4% H_2 for 15 min. Detailed x-ray studies indicate that CeO_2 films have good out-of-plane and in-plane textures with full-width-half-maximum values of 5.8° and 7.5°, respectively. High temperature *in situ* XRD studies show that the nucleation of CeO_2 films starts at 600 °C and the growth completes within 5 min when heated at 1100 °C. SEM and AFM investigations of CeO_2 films reveal a fairly dense microstructure without cracks and porosity. Highly textured YSZ barrier layers and CeO_2 cap layers were deposited on MOD CeO_2 -buffered Ni–W substrates using rf-magnetron sputtering. Pulsed laser deposition (PLD) was used to grow YBCO films on these substrates. A critical current, J_c , of about 1.5 MA cm⁻² at 77 K and self-field was obtained on YBCO (PLD)/ CeO_2 (sputtered)/YSZ (sputtered)/ CeO_2 (spin-coated)/Ni–W.

1. Introduction

Chemical solution processing techniques have emerged as viable low-cost nonvacuum methods for producing ceramic oxide powders and films [1–3]. The most commonly used solution techniques are (i) sol–gel processes that use 2-methoxyethanol as a reactant and solvent; (ii) hybrid processes that use chelating agents such as acetylacetonate or diethanolamine to reduce alkoxide reactivity and (iii) metal organic decomposition (MOD) techniques that use high-molecular-weight precursors and water insensitive carboxylates, 2-ethylhexanoates, etc [4]. These processes offer many desirable aspects, such as precise control of metal oxide precursor stoichiometry and composition, ease of formation of epitaxial oxides, relatively easy scale-up of the film and low cost. In recent years various rare-earth oxides (RE_2O_3) and rare-earth zirconium oxide ($\text{RE}_2\text{Zr}_2\text{O}_7$) films have

been grown epitaxially on biaxially textured Ni and Ni–W substrates by solution based methods [5–7].

In the rolling-assisted biaxially textured substrates (RABiTS) approach, four-layer architecture of CeO_2 /YSZ/ Y_2O_3 /Ni/Ni–W is used to fabricate long lengths of buffered tapes. The purpose of the buffer layers is to retard oxidation of Ni, to reduce the lattice mismatch between Ni and YBCO and also to prevent diffusion of Ni into YBCO. We have chosen cerium oxide, CeO_2 as a potential buffer layer for this study. CeO_2 has a fluorite CaF_2 structure with a lattice parameter of 5.41 Å and it gives better chemical compatibility with Ni–W substrates and also good lattice matching with YBCO. Thin films of CeO_2 have been grown by various vacuum [8–11] and nonvacuum [12] based deposition techniques on rolled-Ni substrates. In this paper, we describe our successful development of the growth of CeO_2 seed layer on rolled Ni–W substrates by MOD

technique which is different from reported chemical solution deposition techniques as in [12], where sol-gel CeO_2 solution buffers were grown on biaxially textured Ni substrates. By using CeO_2 seed layers, we have eliminated the need for Ni over-layers as mentioned above. The three-layer architecture of $\text{CeO}_2/\text{YSZ}/\text{CeO}_2/\text{Ni-W}$ is utilized to produce short prototype samples of YBCO coated conductor in the present study. The CeO_2 seed layer helps us to grow cube textured YSZ buffers on Ni-W substrates; otherwise it is difficult to grow YSZ directly on Ni or Ni-W substrates using our experimental conditions. Here, the CeO_2 seed layer was grown by a solution process. Both YSZ barrier and CeO_2 cap layers were grown by rf-magnetron sputtering and the YBCO films were grown by pulsed laser deposition (PLD) technique.

2. Experimental details

The MOD precursor solution was prepared in ambient atmosphere. The reagents cerium (III) acetylacetonate ($\text{Ce}(\text{acac})_3 \cdot x\text{H}_2\text{O}$), acetic acid and methanol were used as received from Alfa Aesar. $\text{Ce}(\text{acac})_3 \cdot x\text{H}_2\text{O}$ (2.1872 g, 5 mmol) was dissolved in acetic acid (15 ml) by heating in a hot plate at 60°C for 10 min with continuous stirring. Then 5 ml of methanol (25% in volume of the solution) was added, which helps us to stabilize the solution. The final volume of the solution was adjusted to 20 ml by adding methanol to obtain 0.25 M CeO_2 precursor solution. This solution was then spin coated onto short cube textured Ni-W substrates of $2\text{ cm} \times 1\text{ cm}$ in size at 5000 rpm for 30 s; followed by heat treatment at 1100°C for 15 min in a reducing forming gas atmosphere of Ar-4\%H_2 . The samples were introduced into a pre-heated furnace kept at 1100°C after a 5 min purge with Ar-4\%H_2 gas mixture at room temperature. After 15 min heat treatment at 1100°C , the samples were quenched to room temperature with the same atmosphere. The heating and cooling rates were in the range of $350\text{--}400^\circ\text{C min}^{-1}$.

The CeO_2 films were characterized by using x-ray diffraction (XRD) for phase purity and texture, high temperature XRD for nucleation and growth, scanning electron microscopy (SEM) for homogeneity and microstructure and atomic force microscopy (AFM) for surface roughness analysis. A Philips model XRG3100 diffractometer with $\text{CuK}\alpha$ radiation was used to record the θ - 2θ XRD patterns. The texture analysis was performed using a Picker 4-circle diffractometer. High temperature *in situ* XRD experiments were carried out in a flowing atmosphere of He-4\%H_2 and heating ramp of $400^\circ\text{C min}^{-1}$ on a Scintag PAD X diffractometer with an mBraun linear position sensitive detector (PSD) covering a 8° range centred at $2\theta = 31^\circ$. The microstructure analyses of these samples were performed by using a Hitachi S-4100 field emission SEM and Digital Instruments nanoscope AFM in contact mode.

The superconducting quality of the CeO_2 seed layer was tested with a layer sequence of YBCO/ CeO_2 /YSZ/ CeO_2 /Ni-W. Using rf-magnetron sputtering, a 200 nm thick YSZ layer and a 10 nm thick CeO_2 cap layer were deposited on the CeO_2 seeded Ni-W substrates at 780°C in 10 mTorr of forming gas and 2×10^{-6} Torr pressure of water vapour. The YBCO deposition was done by PLD at 790°C in 120 mTorr oxygen with an average laser energy of 400–410 mJ using

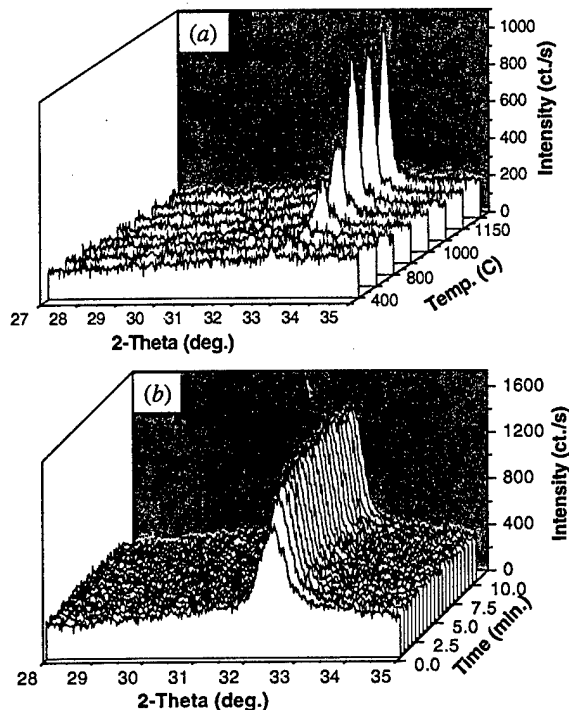


Figure 1. A typical θ - 2θ scan obtained for a 20 nm thick CeO_2 buffered Ni-W substrates in a high temperature *in situ* XRD heat-treated at (a) various temperatures and (b) at a constant temperature of 1100°C for various times. The CeO_2 film has a preferred *c*-axis orientation and the crystallization starts around 600°C (a) and crystallization is completed in 5 min (b).

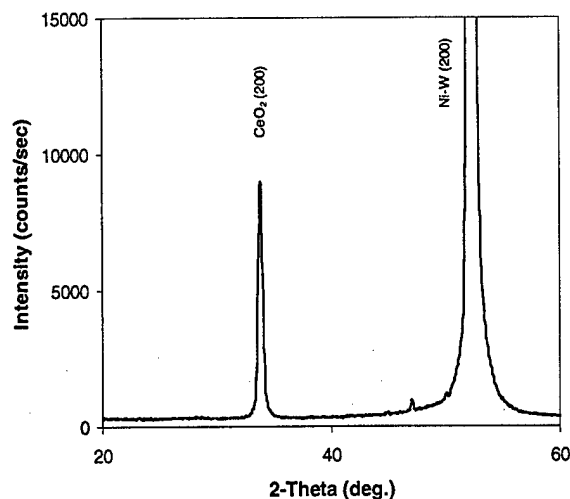


Figure 2. A typical room-temperature θ - 2θ scan obtained for a 20 nm thick CeO_2 buffered Ni-W substrates. The CeO_2 film has a preferred *c*-axis orientation.

a stoichiometric YBCO target, followed by annealing under 550 Torr oxygen during cool down. Details of the PLD process have been published elsewhere [13]. The samples were

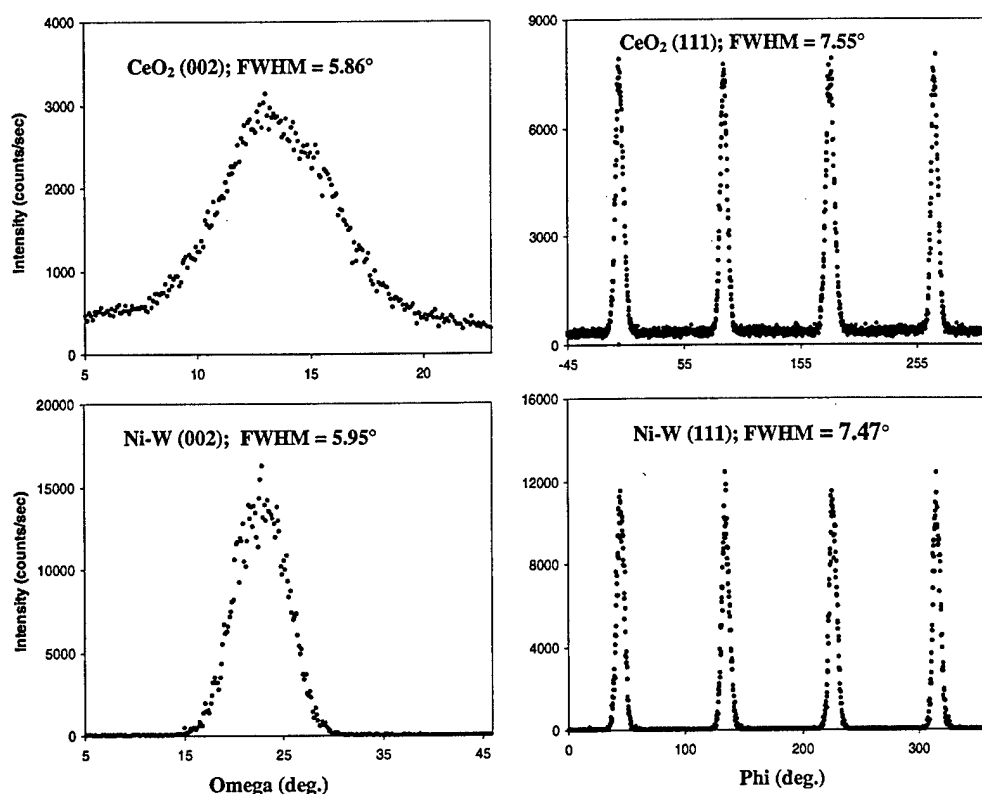


Figure 3. The ω and ϕ scans obtained for a 20 nm thick MOD CeO₂ film grown on textured Ni-W substrate. The FWHM values for each scan are shown inside the patterns.

prepared for electrical property measurements by depositing silver for current and voltage contacts, followed by oxygen annealing at 500 °C for 1 h. The transport critical current density, J_c , was measured using a standard four-point probe technique with an electric-field criterion of 1 $\mu\text{V cm}^{-1}$. The J_c measurements were also performed with the applied magnetic field parallel to the substrate ($H//c$).

3. Results and discussion

3.1. Nucleation and growth analysis by *in situ* HTXRD

The sample was heated from room temperature to 1200 °C at a heating rate of 400 °C min⁻¹ in a reducing atmosphere of He-4%H₂ and the θ -2 θ XRD patterns were recorded at 400, 600, 800, 900, 1000, 1100, 1150 and 1200 °C. For the growth test, a sample was heated to 1100 °C with the same heating ramp and atmosphere and then XRD patterns were recorded for every 30 s for 1 h. Plots for nucleation and growth characteristics of CeO₂ film on textured Ni-W substrates are shown in figures 1(a) and (b). The nucleation of CeO₂ starts at 600 °C and the growth of the film was completed within 5 min when heat-treated at 1100 °C.

3.2. Structure and texture analysis by XRD

A typical θ -2 θ XRD scan for a spin-coated CeO₂ film on the Ni-W substrate is shown in figure 2. The intense CeO₂

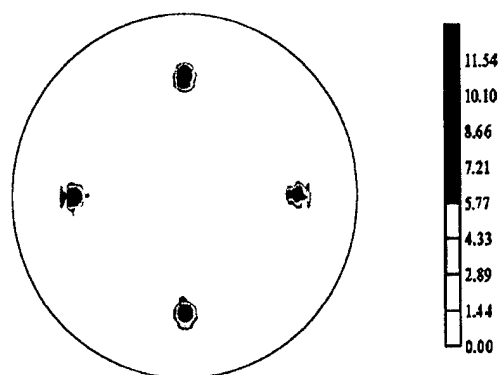


Figure 4. The typical CeO₂ (111) pole figure obtained for a 20 nm thick MOD CeO₂ film grown on the textured Ni-W substrate.

(200) peak reveals the presence of a *c*-axis-aligned film. The ω (out-of-plane) and ϕ (in-plane) scans of these films on the Ni-W substrates are shown in figure 3. The CeO₂ film has a good out-of-plane and in-plane texture with full-width-at-half-maximum (FWHM) of 5.86° and 7.55°, respectively. These values are well comparable to those of Ni-W substrates ($\Delta\omega = 5.95^\circ$, $\Delta\phi = 7.47^\circ$). The typical (111) pole figure for a CeO₂ film grown on the Ni-W substrate is shown in figure 4, which indicates the presence of a single cube-on-cube texture.

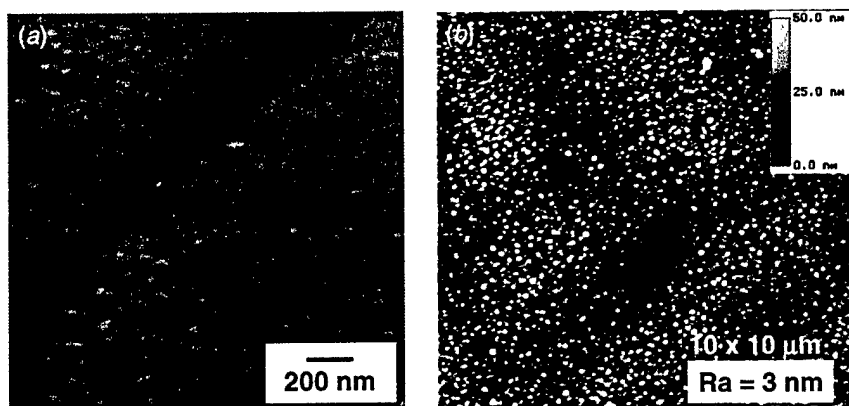


Figure 5. SEM micrograph (a) and AFM image (b) obtained on a 20 nm thick CeO_2 surface.

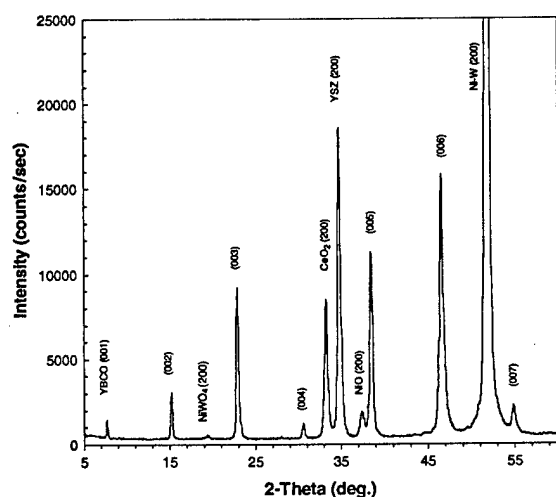


Figure 6. A typical room-temperature θ - 2θ scan obtained for a 200 nm thick PLD YBCO film on a CeO_2 (sputtered)/YSZ (sputtered)/ CeO_2 (MOD)/Ni-W substrate. The YBCO film has a preferred c -axis orientation.

3.3. Microstructure analysis by SEM and AFM

SEM studies of the CeO_2 films exhibit a uniform, smooth and crack free surface morphology as can be observed from figure 5(a). An AFM image of the same sample is shown in figure 5(b). It reveals a root-mean-square roughness (R_a) of the CeO_2 films as 3 nm, which is comparable to that of the underlying alloy substrate ($R_a = 1$ nm). It should be mentioned that for lower concentration of CeO_2 solution (i.e. thinner film) there was a partial coverage of the substrate surface and for thicker CeO_2 films (i.e. higher concentration) crack formation was detected due to lattice mismatch and/or thermal expansion coefficient difference between the CeO_2 and the Ni-W substrate. Details of this observation need further investigation.

3.4. Superconducting test structure

The XRD pattern obtained from the YBCO/ CeO_2 /YSZ/ CeO_2 /Ni-W multilayer structure is shown in figure 6,

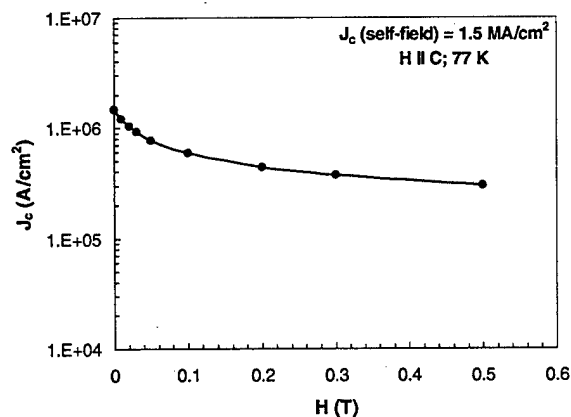


Figure 7. Field dependence of critical current density, J_c , for a 200 nm thick PLD YBCO film on a CeO_2 (sputtered)/YSZ (sputtered)/ CeO_2 (MOD)/Ni-W substrate.

indicating that the YBCO film has a strong c -axis texture. It also reveals the presence of a small amount of NiO and NiWO_4 at the buffer-substrate interface. The magnetic field dependence of J_c (77 K) for a 0.2 μm thick YBCO film grown epitaxially on CeO_2 /YSZ/ CeO_2 /Ni-W is shown in figure 7, where the magnetic field is applied parallel to the c -axis. The highest J_c obtained is about 1.5 MA cm^{-2} for a 0.2 μm thick YBCO film in self-field. The J_c at 0.5 T was about 20% of the zero-field J_c , which resembles the field dependence of a typical YBCO coated conductor. Efforts are being made to optimize the growth of CeO_2 as a cap layer on suitable barrier layers.

4. Conclusions

We have successfully developed a new MOD process to grow epitaxial CeO_2 buffer layers on Ni-W (100) substrates. The spin-coated buffers on Ni-W substrates were smooth, crack free and dense. On spin-coated CeO_2 seed layers, having a total buffer layer sequence of CeO_2 (sputtered)/YSZ (sputtered)/ CeO_2 (spin-coated)/Ni-W, high-quality YBCO films with J_c values around 1.5 MA cm^{-2} at 77 K and self-field

were obtained. By this demonstration, we have eliminated the need for Ni over-layers.

Acknowledgments

Thanks are due to Lee Heatherly for annealing the substrates and Don Kroeger for useful discussions. This work was sponsored by the United States Department of Energy, Office of Energy Efficiency and Renewable Energy, Office of Distributed Energy and Electric Reliability—Superconductivity Program. This research was performed at the Oak Ridge National Laboratory, managed by UT-Battelle, LLC for the USDOE under contract DE-AC05-00OR22725. M S Bhuiyan would also like to acknowledge the help of AFOSR for providing financial support.

References

- [1] Rupich M W, Liu Y P and Ibechem J 1992 *Appl. Phys. Lett.* **60** 1384
- [2] Paranthaman M and Beach D B 1995 *J. Am. Ceram. Soc.* **78** 2551
- [3] Lange F F 1996 *Science* **273** 903
- [4] Schwartz R W 1997 *Chem. Mater.* **9** 2325
- [5] Paranthaman M 2001 *J. Am. Ceram. Soc.* **84** 273
- [6] Sathyamurthy S, Paranthaman M, Aytug T, Kang B W, Martin P M, Goyal A, Kroeger D M and Christen D K 2002 *J. Mater. Res.* **17** 1543
- [7] Aytug T 2003 *J. Am. Ceram. Soc.* **86** 257
- [8] Wu X D, Dye R C, Muenchausen R E, Foltyn S R, Maley M, Rollet A D, Garcia A R and Nogar N S 1991 *Appl. Phys. Lett.* **58** 2165
- [9] Norton D P *et al* 1996 *Science* **274** 755
- [10] Paranthaman M *Physica C* **275** 266
- [11] Varesi E, Celentano G, Petrisor T, Boffa V, Ciontea L, Galluzzi V, Gamberdella U, Mancini A, Rufoloni A and Vannozzi A 2003 *Supercond. Sci. Technol.* **16** 498
- [12] Celik E, Schwartz J, Avci E, Schneider-Muntau H J and Hascicek Y S 1999 *IEEE Trans. Appl. Supercond.* **9** 2264
- [13] Mathis J E, Goyal A, Lee D F, List F A, Paranthaman M, Christen D K, Specht E D, Kroeger D M and Martin P M 1998 *Japan. J. Appl. Phys.* **37** L1379

Growth of Epitaxial Y_2O_3 Film on Biaxially Textured Ni-W Substrates

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ABSTRACT

We have grown epitaxial Y_2O_3 buffer layers on biaxially textured Ni-W substrates using a newly developed metal organic decomposition (MOD) approach. Precursor solution of 0.25M concentration was spin coated on short samples of Ni-3 at.%W (Ni-W) substrates and heat-treated at 1150°C in a gas mixture of Ar-4%H₂ for an hour. Detailed X-Ray studies indicate that Y_2O_3 films has good out-of-plane and in-plane textures with full-width-half-maximum values of 6.22° and 7.51°, respectively. SEM investigations of Y_2O_3 films reveal a fairly dense microstructure without cracks and porosity. It is possible to use this MOD Y_2O_3 template for growing high current density YBCO films.

INTRODUCTION

Chemical solution processing techniques have emerged as viable low-cost nonvacuum methods for producing ceramic oxide powders and films [1-3]. These processes offer many desirable aspects, such as precise control of metal oxide precursor stoichiometry and composition, ease of formation of epitaxial oxides, relatively easy scale-up of the film and low cost. In recent years various rare-earth oxides (RE_2O_3) and rare-earth zirconium oxide ($\text{RE}_2\text{Zr}_2\text{O}_7$) films have been grown epitaxially on biaxially textured Ni and Ni-W substrates by solution based methods [4-7]. In the Rolling-Assisted Bi-axially Textured Substrates (RABiTS) approach, four-layer architecture of $\text{CeO}_2/\text{YSZ}/\text{Y}_2\text{O}_3/\text{Ni}/\text{Ni-W}$ is used to fabricate long lengths of buffered tapes. The purpose of the buffer layers is to retard oxidation of Ni, to reduce the lattice mismatch between Ni and YBCO and also to prevent diffusion of Ni into YBCO. We have chosen Ytria, Y_2O_3 as a potential buffer layer for this study. Y_2O_3 has a bcc structure with a lattice parameter of 10.6 Å and it has a good chemical compatibility with Ni-W and also good thermal stability. Thin films of Y_2O_3 have been grown by various vacuum [8,9] based deposition techniques on rolled-Ni substrates, whereas no non-vacuum technique for deposition of Y_2O_3 has been reported. In this paper, we describe our successful development of the growth of Y_2O_3 buffer layer on rolled Ni-W substrates by MOD technique.

EXPERIMENTAL DETAILS

The MOD precursor solution was prepared in ambient atmosphere. The reagents yttrium (III) acetylacetonate ($\text{Y}(\text{acac})_3 \cdot x\text{H}_2\text{O}$), acetic acid and methanol were used as received from Alfa Aesar. $\text{Y}(\text{acac})_3 \cdot x\text{H}_2\text{O}$ (1.93 g, 5 mmol) was dissolved in acetic acid (15 ml) by heating in a hot plate at 60 °C for 10 minutes with continuous stirring. Then 5 ml of methanol (25% in volume of the solution) was added, which helps to stabilize the solution. The final volume of the solution was adjusted to 20 ml by adding methanol to obtain 0.25 M Y_2O_3 precursor solution. This solution was then spin coated onto short cube textured Ni-W substrates of 2 cm x 1 cm in size at

5000 rpm for 30 sec; followed by heat treatment at 1150 °C for 1 h in a reducing atmosphere of Ar-4% H₂. The samples were introduced into a pre-heated furnace kept at 1150 °C after a 5 minutes purge with Ar-4% H₂ gas mixture at room temperature. After 1 h heat-treatment at 1150 °C, the samples were quenched to room temperature with the same atmosphere.

The Y₂O₃ films were characterized by using x-ray diffraction (XRD) for phase purity and texture and scanning electron microscopy (SEM) for homogeneity and microstructure. A Philips model XRG3100 diffractometer with Cu K α radiation was used to record the θ -2 θ XRD patterns. The texture analysis was performed using a Picker 4-circle diffractometer. Using a Hitachi S-4100 field emission SEM performed the microstructure analyses of these samples.

DISCUSSION

The θ -2 θ XRD scan for a spin-coated Y₂O₃ film on Ni-W substrate has revealed the presence of a c-axis-aligned film. The typical (222) pole figure for a Y₂O₃ film grown on the Ni-W substrate is shown in Fig. 1, which indicates the presence of a single cube-on-cube texture. The Y₂O₃ film has a good out-of-plane and in-plane texture with full-width-at-half-maximum (FWHM) of 6.22° and 7.51°, respectively. These values are well comparable to those of Ni-W substrates ($\Delta\omega = 5.04^\circ$, $\Delta\phi = 7.33^\circ$). SEM micrographs for Y₂O₃ buffer layers on rolled Ni-W substrate using spin coating are shown in Fig. 2. As seen from figure 2, Y₂O₃ buffer layers provide very good coverage for the Ni-W surface. Most of the Ni-W grain boundary grooves on the Ni-W surface were found to be well covered. Figure 2 also shows that the buffer layers are continuous as well as crack free. These results indicate MOD techniques can produce continuous, dense and crack-free buffer layers on rolled Ni-W substrate. Efforts are being made to deposit sputtered YSZ and CeO₂ on MOD Y₂O₃/Ni-W substrate and YBCO by pulsed laser deposition (PLD) on CeO₂/YSZ/Y₂O₃ (MOD)/Ni-W.

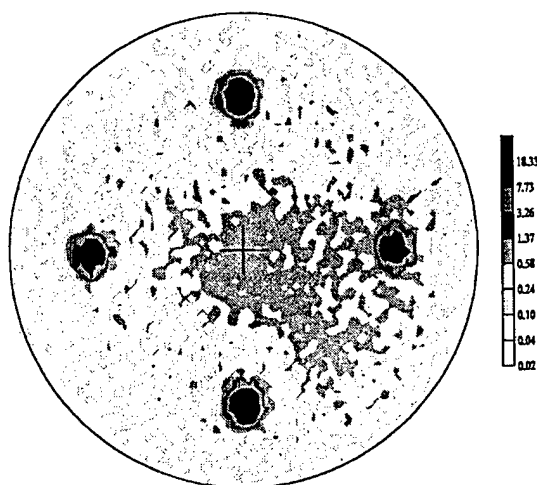


Fig.1. Y₂O₃ (222) pole figure on log scale

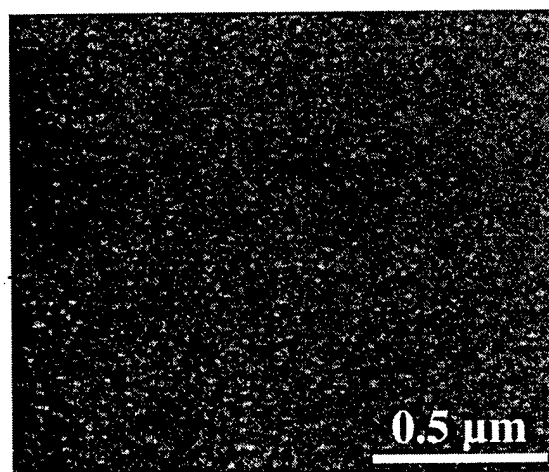


Fig.2. SEM micrographs on a 15 nm thick Y₂O₃ surface.

CONCLUSION

We have successfully developed a new MOD process to grow epitaxial Y_2O_3 buffer layers on Ni-W (100) substrates. In-plane and out-of-plane alignments indicate the buffer layers are sharp textured. Pole figures of buffer layers show a predominantly single-cube texture. SEM reveals a continuous, dense and crack-free microstructure for MOD-coated buffers which indicates that the surface of Y_2O_3 buffer layers deposited on rolled Ni substrates using MOD process is suitable for further deposition of other buffer layers.

ACKNOWLEDGEMENTS

Thanks are due to Lee Heatherly for annealing the substrates and Don Kroeger for useful discussion. This work was sponsored by the United States Department of Energy, Office of Electric Transmission and Distribution. This research was performed at the Oak Ridge National Laboratory, managed by U.T.-Battelle, LLC for the USDOE under contract DE-AC05-00OR22725. M. S. Bhuiyan would also like to acknowledge the help of AFOSR for providing a financial support.

REFERENCES

- [1] Rupich M W, Liu Y P and Ibechem J 1992 *Appl. Phys. Lett.* **60** 1384
- [2] Paranthaman M and Beach D B 1995 *J. Am. Ceram. Soc.* **78** 2551
- [3] Lange F F 1996 *Science* **273** 903
- [4] Paranthaman M, Chirayil T G, List F, Cui X, Goyal A, Lee D F, Specht E D, Martin P M, Williams R K, Kroeger D M, Morrel J S, Beach D B, Feenstra R and Christen D K 2001 *J. Am. Ceram. Soc.* **84** 273
- [5] Sathyamurthy S, Paranthaman M, Aytug T, Kang B W, Martin P M, Goyal A, Kroeger D M and Christen D K 2002 *J. Mat. Res.* **17** 1543
- [6] Aytug T, Paranthaman M, Kang B W, Beach D B, Sathyamurthy S, Specht E D, Lee D F, Feenstra R, Goyal A, Kroeger D M, Leonard k J, Martin P M and Christen D K 2003 *J Am. Ceram. Soc.* **86** 257
- [7] Bhuiyan M S, Paranthaman M, Sathyamurthy S, Aytug T, Kang S, Lee D F, Goyal A, Payzant E A and Salama K, 2003 *Supercond. Sci. Technol.* **16** 1305
- [8] Paranthaman M, Lee D F, Goyal A, Specht E D, Martin P M, Cui X, Mathis J E, Feenstra R, Christen D K and Kroeger D M, 1999 *Supercond. Sci. Technol.* **12** 319
- [9] Ivanic R, Rehacek V, Novotny I, Breternitz V, Spiess L, Knedlik C, Tvarozek V 2001 *Vacuum* **61** 229

EPITAXIAL GROWTH OF SOLUTION BASED RARE EARTH NIOBATE, RE_3NbO_7 FILMS ON BIAXIALLY TEXTURED Ni-W SUBSTRATES

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A new series of single rare earth niobate, RE_3NbO_7 ($\text{RE}=\text{Y}, \text{Gd}, \text{Eu}$) buffer layers have been developed for the growth of superconducting $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ (YBCO) films on biaxially textured Ni-W (3 at.%) substrates. Using a metal-organic deposition (MOD) process, smooth, crack-free and epitaxial RE_3NbO_7 ($\text{RE}=\text{Y}, \text{Gd}, \text{Eu}$) films were grown on cube textured Ni-W substrate. YBCO film with a critical current density of 1.1×10^6 A/cm² in self-field at 77 K has been grown on Gd_3NbO_7 -buffered Ni-W substrate using pulsed laser deposition. This demonstration promises a route for producing low-cost all solution buffers for second generation YBCO coated conductors.

The main objective of this work is to research and develop faster, potentially lower-cost, and simpler RABiTS architectures as alternate to the standard three-layer architecture of YBCO/CeO₂/YSZ/Y₂O₃/Ni-W. During the search for new innovative buffer layers, we started exploring rare earth niobium oxides as the potential candidates. As per the phase diagram of the Y₂O₃-Nb₂O₅ system, YNbO₄ and Y₃NbO₇ (pyrochlore: Y₂(YNb)O₇) phases are known to exist. Both phases may be used as the buffer layers. In the present work, we have studied the Y₃NbO₇ system. Y₃NbO₇ is a cubic pyrochlore with a lattice parameter of 5.2499 Å (pseudo cubic lattice parameter: 3.713 Å). Similarly, RE₃NbO₇ (RE=Rare Earth) phases are also known in the literature. Since chemical solution deposition has been emerged as a viable, low-cost, non-vacuum process for fabricating long lengths of YBCO coated conductors, we have selected the chemical precursor route to prepare RE₃NbO₇ films. RE₃NbO₇ films can be prepared by both sol-gel alkoxide precursor route and metal-organic deposition (MOD). Here, we report our successful demonstration of the growth of single RE₃NbO₇ (RE=Y, Eu and Gd) solution buffer on strengthened and textured Ni-W (3 at. %) substrates.

The starting reagents were weighed in an argon-filled, inert-atmosphere glove box, and the solution preparation was carried out under argon, with a standard Schlenk-type apparatus. Yttrium (III) isopropoxide (Alfa, 92-95%), Niobium ethoxide (Alfa, 99.999%), and 2-methoxyethanol (Alfa, spectrophotometric grade) were used without further purification. For preparing Eu₃NbO₇ and Gd₃NbO₇, Europium (III) acetate (Alfa, 99.9%) and Gadolinium (III) acetate (Alfa, 99.9%) were used after purification. The Y₃NbO₇ (YNO) precursor solution with 0.4 M total cation concentration was prepared by refluxing stoichiometric quantities of Yttrium isopropoxide and Niobium ethoxide in 2-methoxyethanol. The YNO precursor solution was then spin-coated onto short coupons of cube textured Ni-W substrates of 2 cm x 1 cm in size at 5000 rpm for 30 s; followed by heat treatment at 1100 °C for 15 min in a flowing reducing atmosphere of Ar-4%H₂. The tapes were introduced into a pre-heated furnace kept at 1100 °C after a 5 min purge with Ar-4%H₂ gas mixture at room temperature. After 15 min of heat treatment at 1100 °C, the tapes were then quenched to room temperature in the same atmosphere. Similarly,

Eu_3NbO_7 (ENO) and Gd_3NbO_7 (GNO) films on textured Ni-W substrates were prepared at 1050 °C/15 min and 1100 °C/15 min, respectively. About 20 nm thick RE_3NbO_7 films were produced in a single coat. Multiple coatings were made to prepare thick films. On the Gd_3NbO_7 -buffered Ni-W tape, YBCO films were deposited using pulsed laser deposition (PLD) at 780 °C in 120mTorr oxygen with average laser energy of 200 mJ.

The samples were characterized for phase purity and texture using XRD, and the microstructure of both RE_3NbO_7 (RE=Y, Eu, Gd) and YBCO were monitored using scanning electron microscope (SEM). The resistivity and transport critical current density, J_c , were measured using a standard four-point probe technique. The voltage contact spacing was 0.4 cm. Values of J_c were calculated using a 1 $\mu\text{V}/\text{cm}$ criterion. Electrical contacts of Ag were deposited onto the samples using dc sputtering followed by an O_2 annealing in 1 atm for 30 minutes at 500 °C.

Typical θ - 2θ XRD pattern of a single coat RE_3NbO_7 (RE=Y, Eu, Gd) film on a textured Ni-W substrate is shown in Figure 1. These scans indicate the presence of a strong c-axis (200) aligned films. Similar results were obtained for 3-4 coats of Gd_3NbO_7 and Eu_3NbO_7 films on Ni-W substrates. Thickness of the film is estimated to be 20 nm/coat. The ω and ϕ scans for a 20 nm thick Gd_3NbO_7 film on a textured Ni-W substrate is reported in Figure 2. The FWHM values for Gd_3NbO_7 (002) and Ni-W (002) were 7.15°, and 5.51° respectively. Similarly, the FWHM values for Gd_3NbO_7 (111) and Ni-W (111) were 8.0°, and 6.72° respectively. The (111) X-ray pole figures for 20 nm thick RE_3NbO_7 (RE=Y, Eu, Gd) buffers on Ni-W substrate are shown in Figure 3. The presence of a four-fold symmetry indicates the presence of a single cube textured RE_3NbO_7 film. As shown in Figure 4, crack-free RE_3NbO_7 (RE=Y, Eu, Gd) buffers were produced. As shown in Figure 5, the θ - 2θ XRD pattern of a YBCO film on Gd_3NbO_7 -buffered Ni-W substrate indicated the presence of a strong (200) reflection of YBCO films. The transport properties for the YBCO film grown on Gd_3NbO_7 (3-coats) buffered Ni-W substrate is shown in Figure 6. The zero field J_c of over $1.1 \times 10^6 \text{ A}/\text{cm}^2$ at 77 K and self-field were obtained.

In a summary, epitaxial RE_3NbO_7 ($\text{RE}=\text{Y}, \text{Gd}, \text{Eu}$) films have been deposited on textured Ni-W substrates using a solution deposition. Gd_3NbO_7 films have been used as a single buffer layer for deposition of YBCO by PLD on these metal substrates. Critical current densities of over 1.1 MA/cm^2 have been measured for the Ni-W substrates. The present invention offers promise to long length coated-conductor fabrication using a single buffer layer processed using a scaleable solution deposition route.

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REFERENCES

FIGURE CAPTIONS

FIGURE 1. A typical θ - 2θ scan for a 20 nm thick RE_3NbO_7 ($\text{RE}=\text{Y}, \text{Gd}, \text{Eu}$) film grown on biaxially textured Ni-W substrates using metal-organic deposition. RE_3NbO_7 film has a preferred c-axis orientation.

FIGURE 2. The ω and ϕ scans for a 20 nm thick Gd_3NbO_7 film grown on biaxially textured Ni-W substrates using metal-organic deposition. The FWHM values for each scan are shown inside the scans.

FIGURE 3. The (111) pole figure for 20 nm thick (a) Y_3NbO_7 , (b) Eu_3NbO_7 , and (c) Gd_3NbO_7 .

FIGURE 4. SEM micrograph for a 20 nm thick (a) Y_3NbO_7 , (b) Eu_3NbO_7 , and (c) Gd_3NbO_7 film surface.

FIGURE 5. A typical θ - 2θ scan for a 200 nm thick YBCO film grown on 3-coats of 60 nm thick Gd_3NbO_7 buffered Ni-W substrates using pulsed laser deposition. YBCO film has a preferred c-axis orientation. In addition, small amounts of NiO impurities and polycrystalline YBCO peaks were identified.

FIGURE 6. Superconducting properties of a 200 nm thick YBCO film grown on Gd_3NbO_7 buffered Ni-W substrates using pulsed laser deposition. The self-field J_c of 1.1 MA/cm² at 77 K was obtained.

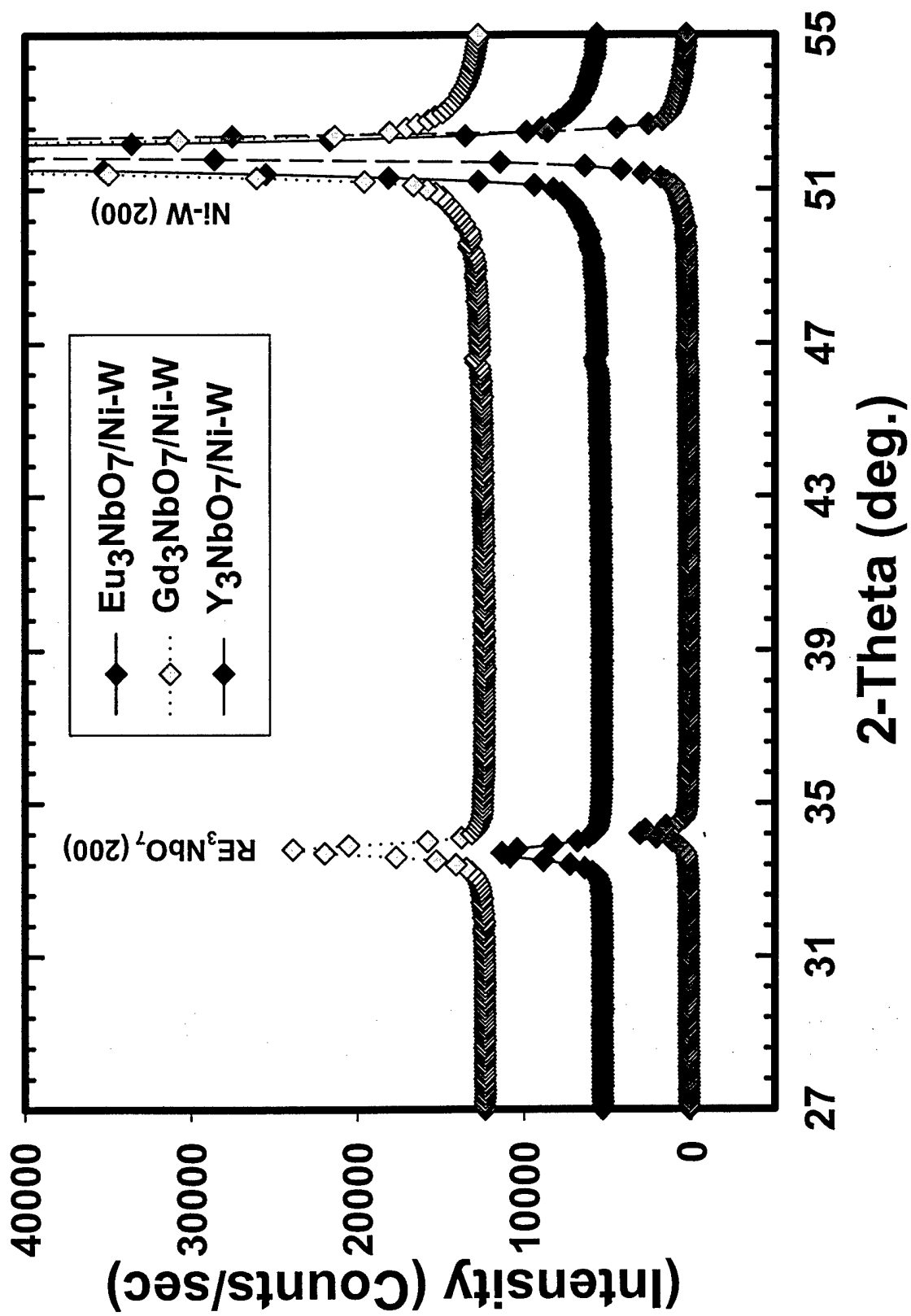


Figure 1 Paranthaman et al.

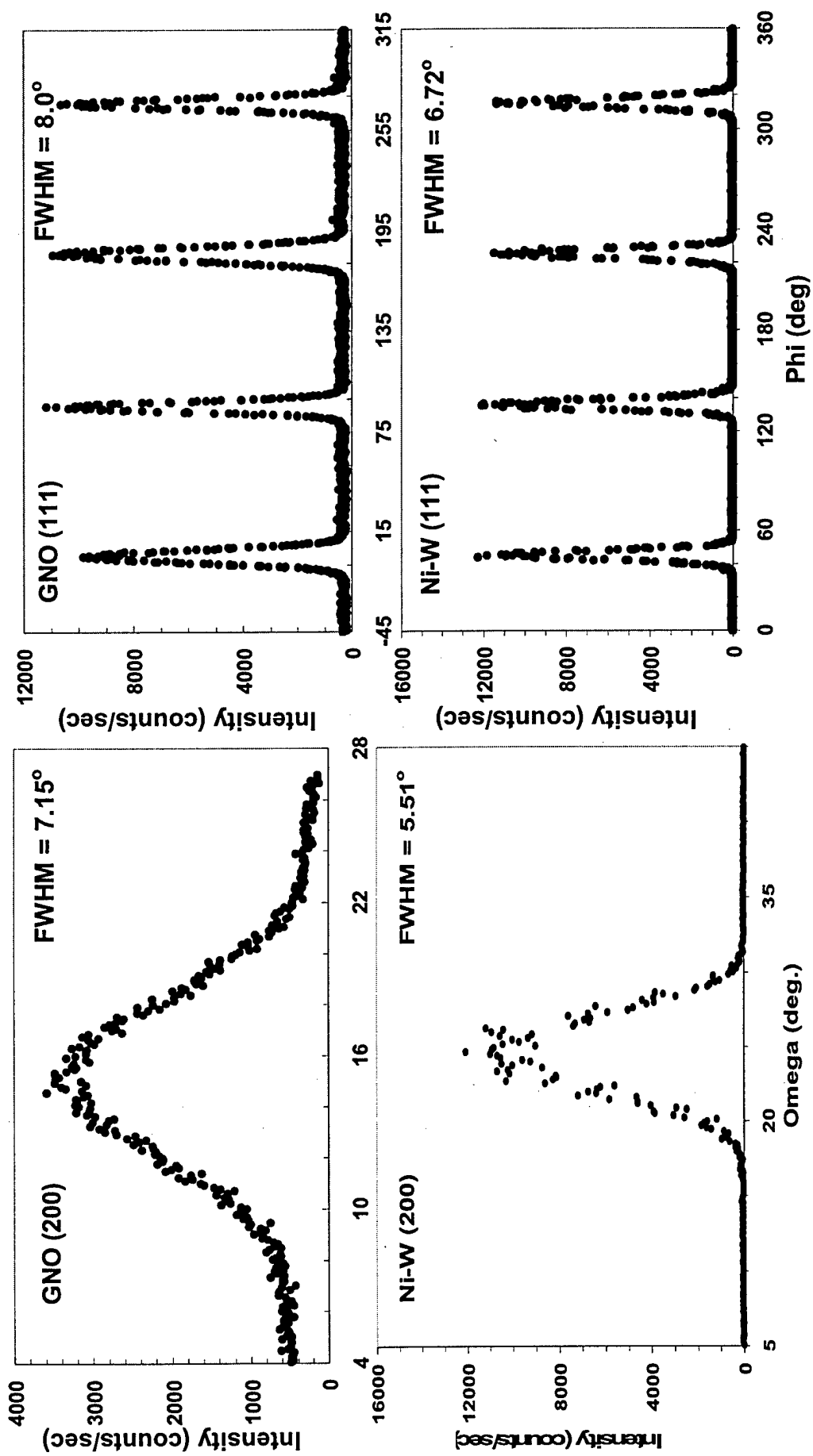
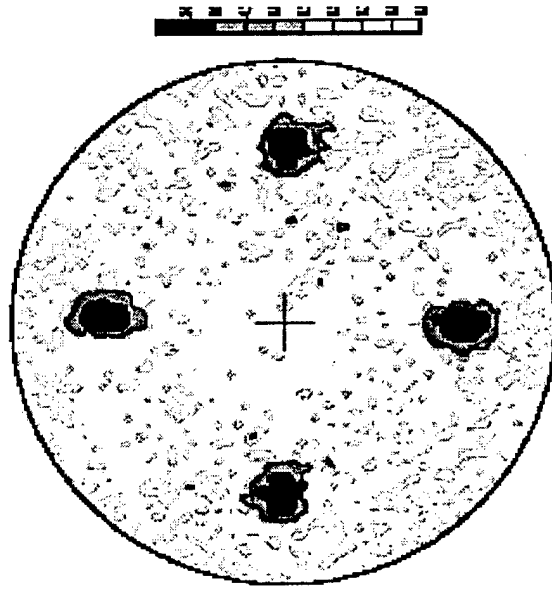
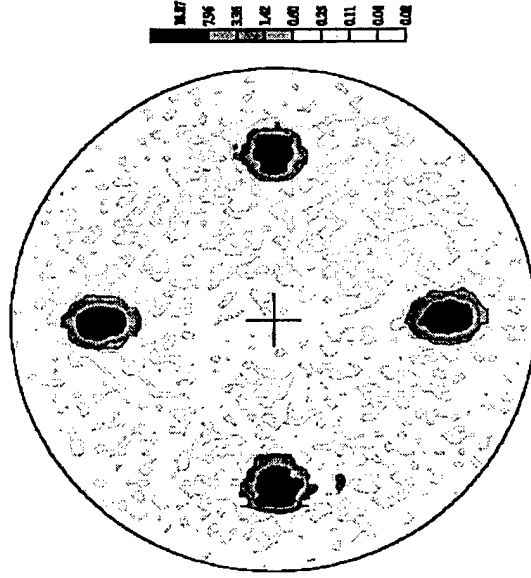


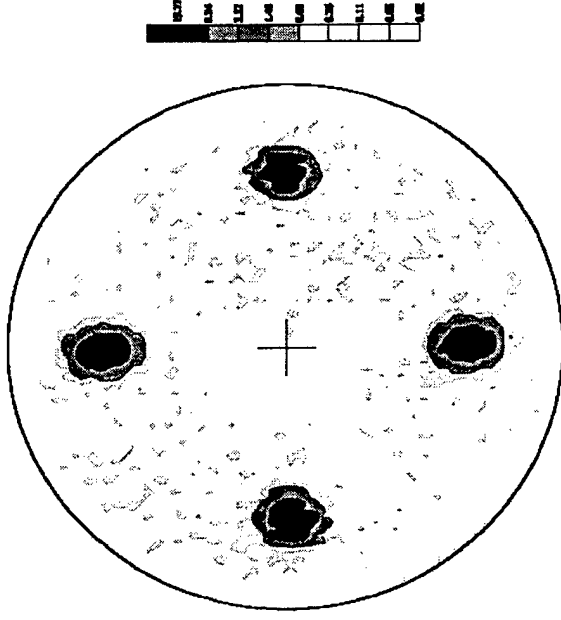
Figure 2 Paranthaman et al.



YNO (111)
(a)



ENO (111)
(b)



GNO (111)
(c)

Figure 3 Paranthaman et al.

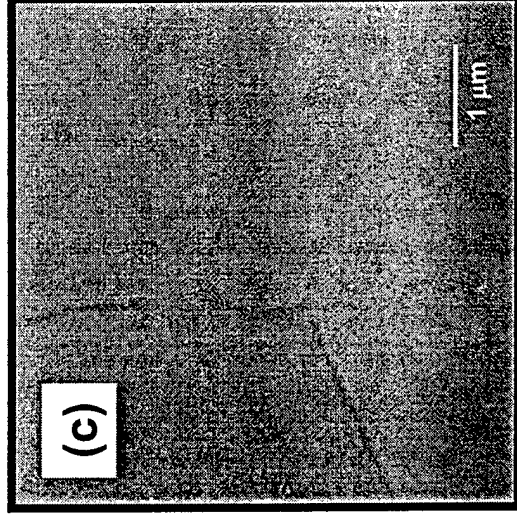
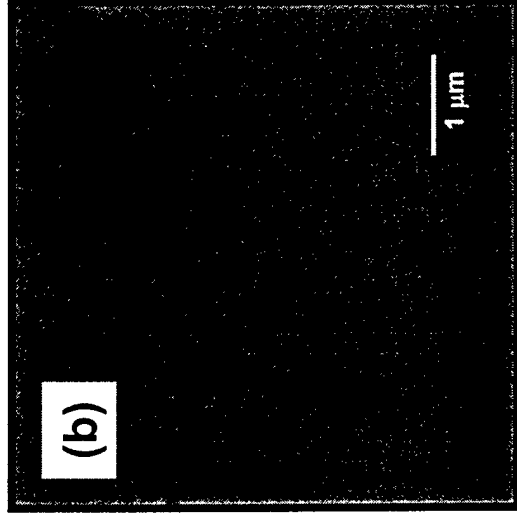
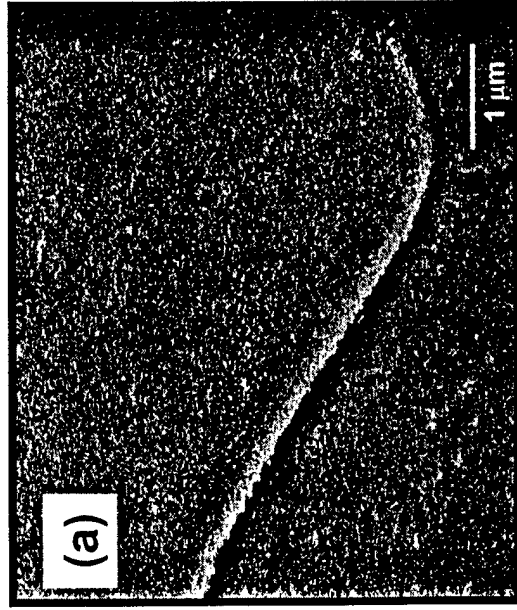


Figure 4 Paranthaman et al.

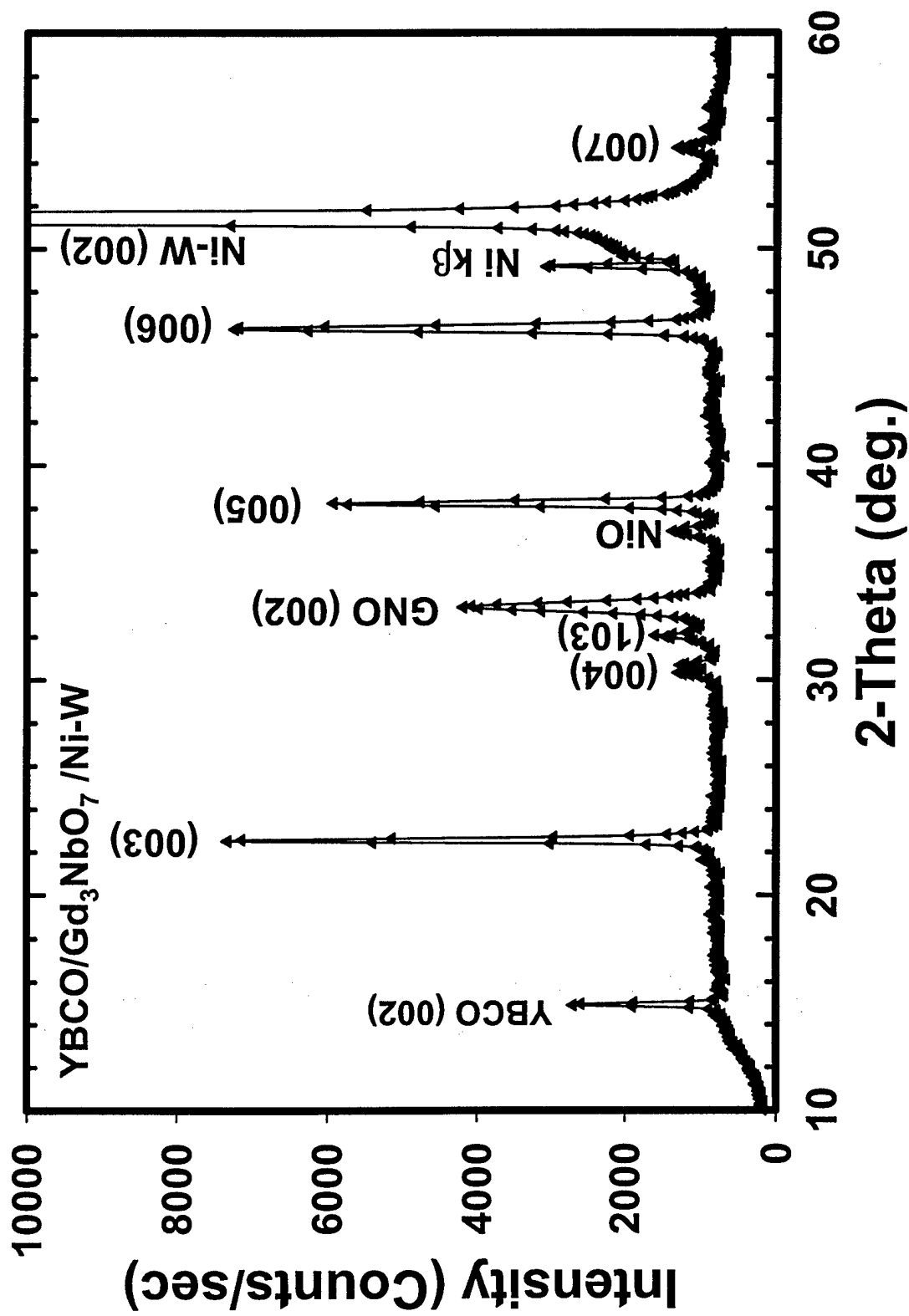


Figure 5 Paranthaman et al.

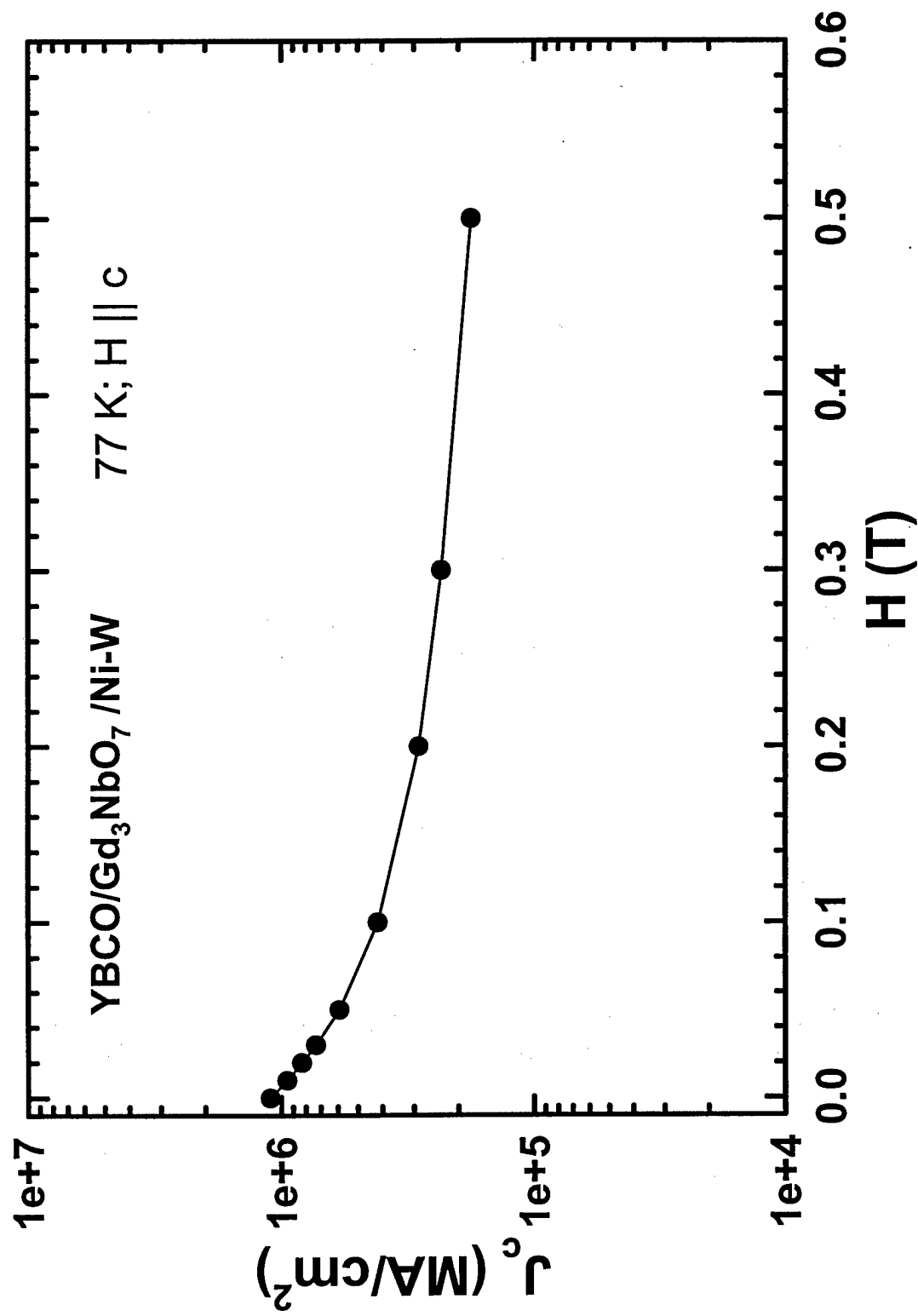


Figure 6 Paranthaman et al.